



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/GB97/02275 <b>(22) International Filing Date:</b> 26 August 1997 (26.08.97) <b>(30) Priority Data:</b> 9618377.7 3 September 1996 (03.09.96) GB <b>(71) Applicant (for all designated States except US):</b> FLEXITAL- LIC LIMITED [GB/GB]; P.O. Box 21, Rochdale OL12 7EU (GB). <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only):</b> HOYES, John, Robert [GB/GB]; 2 The Brook, Calderbrook Road, Littleborough OL15 9NW (GB). STANCLIFFE, Alan [GB/GB]; 87 High- croft Way, Syke, Rochdale, Lancashire OL12 9UE (GB). <b>(74) Agents:</b> SHERRARD-SMITH, Hugh et al.; Appleyard Lees, 15 Clare Road, Halifax, West Yorkshire HX1 2HY (GB).		<b>(81) Designated States:</b> AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, HU, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
<b>(54) Title:</b> STRENGTHENED GRAPHITE SHEET MATERIAL  <b>(57) Abstract</b>  A strengthened exfoliated graphite sheet material and a method for making same is described. The method comprises the steps of applying at least one coat of a liquid plastics material to at least one face of said exfoliated graphite sheet material and drying said at least one liquid coat to leave an adherent plastics material sheet on said graphite sheet material.		

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STRENGTHENED GRAPHITE SHEET MATERIAL

The present invention relates to graphite sheet material having improved strength and handlability.

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Exfoliated or expanded graphite may be compressed to form sheet material which finds many applications in sealing gaskets for example owing to its high compressibility and conformability. However, exfoliated graphite sheet is relatively very brittle, easily fracturing when bent or roughly handled. Various exfoliated graphite sheet materials having improved strength and durability have been proposed. GB2154675 of common ownership herewith describes an expanded graphite web material having a reinforcing core of thin metal, the material being made by laminating a sheet of expanded graphite to each face of a thin metal, e.g. nickel, sheet by adhesive means. The tensile strength of the resulting graphite sheet is greatly increased. Another proposal has involved the bonding by adhesive means of a plastics material sheet film to one or both faces of a graphite sheet.

Whilst these earlier proposals have resulted in exfoliated graphite sheet having greatly improved strength and durability, they suffer from the disadvantage of being costly to manufacture due, inter alia, to the requirement of needing expensive capital equipment for their production. Furthermore, when a plastics sheet is bonded to graphite sheet the sheet must be at a minimum thickness to enable the sheet to be passed through rollers and applied to the graphite. Thus when the gasket is clamped between two flanges and subject to heat, as in a cylinder head gasket, the plastics will stick to the flange. This

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The plastics material sheet may be applied in several coats to one or both faces of the graphite sheet material as desired. When more than one coat is applied, each coat may be dried prior to application of the next or  
5 alternatively, more than one coat may be dried at a time so long as the total coating is self supporting without running off the sheet surface for example. The number of coats applied will be dependent upon the final desired thickness and strength of the plastics material sheet and  
10 the desired degree of enhancement of strength of the graphite sheet.

Depending upon the plastics material being applied, it may also be necessary to further include a curing step  
15 in addition to or as part of the drying step. The curing step may be to polymerise the dried coating and increase the strength of the plastics material coating itself. The temperature at which drying and/or the curing steps will be carried out will be dependent upon the type of plastics  
20 material in question.

Where more than one coat of plastics material is applied these may not necessarily all be of the same material.  
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The thickness of each individual plastics material layer may be controlled to an extent by the viscosity of the applied liquid coat, the greater the viscosity, the thicker each coat may be.  
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An example of a suitable plastics material may be a water based acrylic polymer resin.

Two or more coats of plastics material may be  
35 applied. It has been found that depending upon the

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equal to 10:1 or less than or equal to 5:1 or in the region of 4:1.

Strengthened exfoliated graphite sheet according to the present invention is suitable for the manufacture of so-called spiral wound gaskets and also for the protection of already formed spiral wound gaskets by coating thereof. The plastics material coating prevents the edges of exfoliated graphite sheet material from being friable and easily chipping, either during cutting or during subsequent handling and storage.

The surface of the graphite material itself may be treated for example by indenting or shallow spiking or by chemical means so as to improve the mechanical bonding of the resulting plastics material to the surface.

In order that the present invention may be more fully understood, examples will now be described by way of illustration only.

Exfoliated graphite sheet of thickness about 1mm was selected for testing. This material would fracture across its width when bent through a relatively small angle of about 10 to 20°.

Three different self-crosslinking acrylic resin emulsions were chosen for their differing physical characteristics. These were :

30      Revacryl 274 (trade name) - soft, flexible Tg -24°C  
Revacryl 275 (trade name) - tough, flexible Tg -5°C  
Revacryl 277 (trade name) - tough, stiff Tg +10°C

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specification, and the contents of all such papers and documents are incorporated herein by reference.

5 All of the features disclosed in this specification (including any accompanying claims, abstract and drawings), and/or all of the steps of any method or process so disclosed, may be combined in any combination, except combinations where at least some of such features and/or steps are mutually exclusive.

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Each feature disclosed in this specification (including any accompanying claims, abstract and drawings), may be replaced by alternative features serving the same, equivalent or similar purpose, unless expressly stated otherwise. Thus, unless expressly stated otherwise, each feature disclosed is one example only of a generic series of equivalent or similar features.

20 The invention is not restricted to the details of the foregoing embodiment(s). The invention extends to any novel one, or any novel combination, of the features disclosed in this specification (including any accompanying claims, abstract and drawings), or to any novel one, or any novel combination, of the steps of any  
25 method or process so disclosed.

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9. A method as claimed in any preceding claim comprising adding filler to the liquid prior to applying the coat of plastics material.
- 5 10. A method of making a strengthened graphite sheet material substantially as hereinbefore described with reference to the accompanying description and examples.
- 10 11. Strengthened exfoliated graphite sheet material, said exfoliated graphite sheet material having on at least one face thereof a sheet of a plastics material applied initially as a liquid.
- 15 12. A strengthened graphite sheet material according to claim 11 wherein said plastics material is a water-based acrylic polymer resin.
- 20 13. A strengthened graphite sheet material according to either claim 11 or claim 12 further including a release agent.
- 25 14. A strengthened graphite sheet material according to claim 13 wherein said release agent is provided as a separate outer layer.
15. A strengthen graphite sheet material according to claim 13 wherein said release agent is incorporated into said plastics material sheet.
- 30 16. A strengt6hene graphite sheet material according to claim 1 or any one of claims 11 to 15 wherein porosity adjacent the surface of the graphite material is impregnated with the plastics material.

## INTERNATIONAL SEARCH REPORT

Int. l. Application No.

PCT/GB 97/02275

A. CLASSIFICATION OF SUBJECT MATTER  
IPC 6 C04B41/83 F16J15/10

According to International Patent Classification(IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C04B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	PATENT ABSTRACTS OF JAPAN vol. 014, no. 342 (C-0743), 24 July 1990 & JP 02 124768 A (HITACHI CHEM CO LTD), 14 May 1990, see abstract	1-5,8, 10,11, 16,20
A	-& DATABASE WPI Section Ch, Week 9025 Derwent Publications Ltd., London, GB; Class E36, AN 90-189654 XP002050298 see abstract	12

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

## \* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"Z" document member of the same patent family

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# INTERNATIONAL SEARCH REPORT

Information on patent family members

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Patent document cited in search report	Publication date	Patent family member(s)	Publication date
DE 4332346 A	30-03-95	NONE	
EP 0519620 A	23-12-92	US 5198063 A DE 69222472 D JP 6024872 A	30-03-93 06-11-97 01-02-94

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